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Photoacoustic Observations of Aerosol Optical Properties Aloft Alaska: Quantifying Arctic Radiative Forcing

Manvendra Dubey (LANL)

Claudio Mazzoleni (MTU, LANL)

Alla Zelenyuk (PNL) and the ISDAC team



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Outline

- **Motivation**

*Models under-predict Arctic warming & ice melting
Arctic aerosol forcing is large and episodic but uncertain*

- **ISDAC Observational Strategy**

*State-of-art in situ measurements of optical properties
(photoacoustic), chemistry/size distributions (SPLAT).*

- **Key Objective**

*Prognostic models for aerosol optical properties from size
distributions and chemical composition (mixing state, coating)*

- **Fresh Results to Guide Modelers**

*Quantify spring forcing by Arctic haze over Alaska
Improve Arctic aerosol-cloud-radiation treatments.*

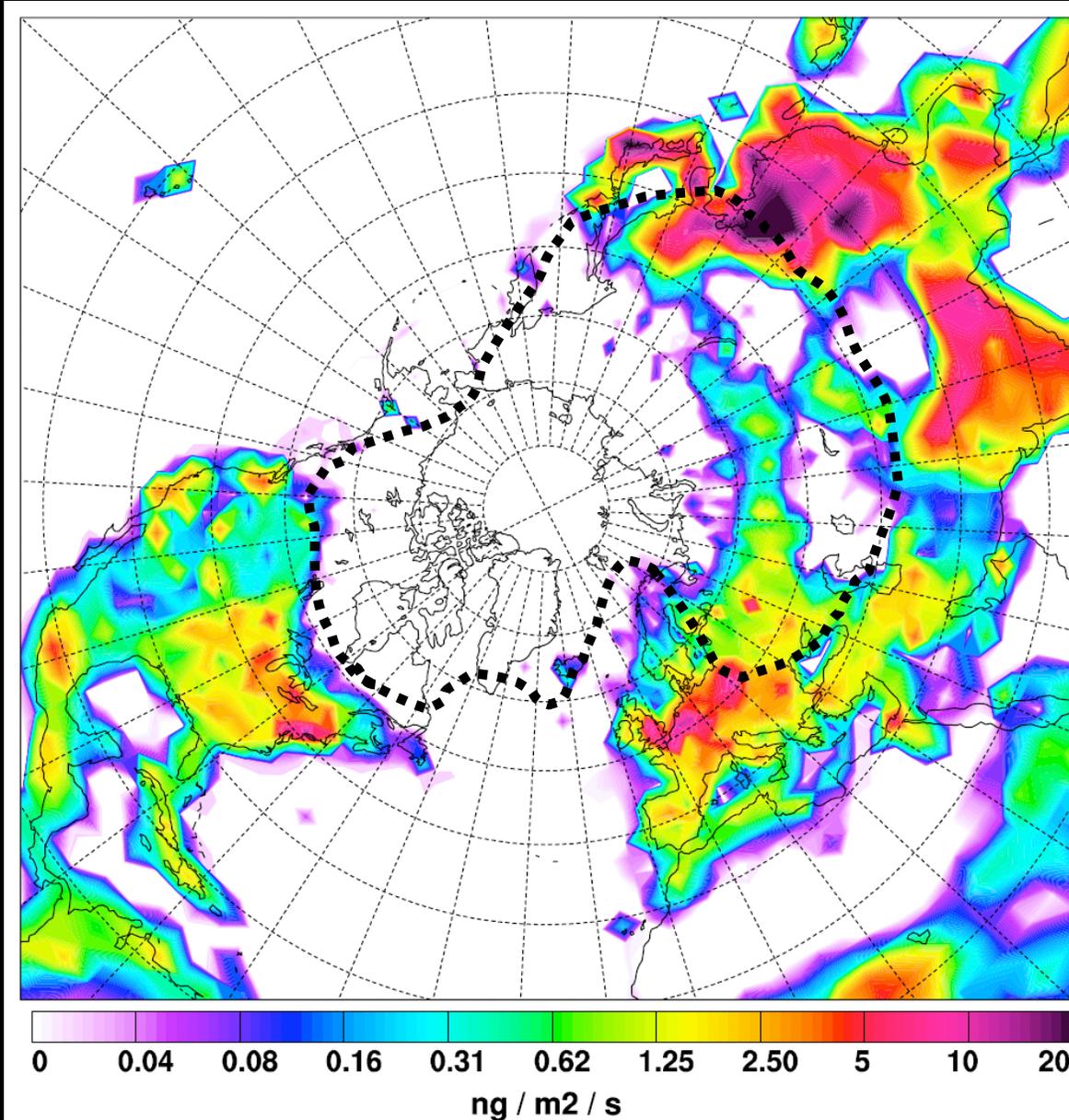
Challenges for Arctic Assessments

- Model transport of long-range pollutants into Arctic
- Capture variability in Arctic dynamics of fires/dust
- Is fossil/biofuel energy the major human addition?
- Is Arctic oil-gas exploration affecting Siberian fires?
- How will NW passage shipping exacerbate pollution?
- Reliable baseline and pollution emissions scenarios.

Anthropogenic sources of soot (industrial, biofuel, fires)

Sources in northern Europe and NE China are consistently within or near the mean position of the Arctic front.

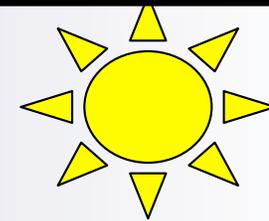
Stohl et al., 2006



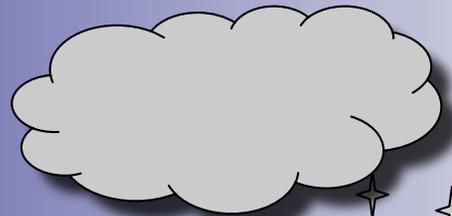
NASA GISS

SPRING: INSTANTANEOUS AEROSOL FORCING

Add Fossil + Biofuel to current Biomass Burning



$$F^{TOA} = 0.9 \text{ W m}^{-2}$$



$$F^{ATM} = 1.6 \text{ W m}^{-2}$$

Aerosol Direct

$$F^S = -0.7 \text{ W m}^{-2}$$

Black carbon
Snow albedo
+ ΔT

$$F^{BC_{snow}} = 0.53 \text{ W m}^{-2}$$

$$\Delta T_s = -0.93 \text{ C}$$

$$\Delta T_s = +0.6 \text{ C}$$

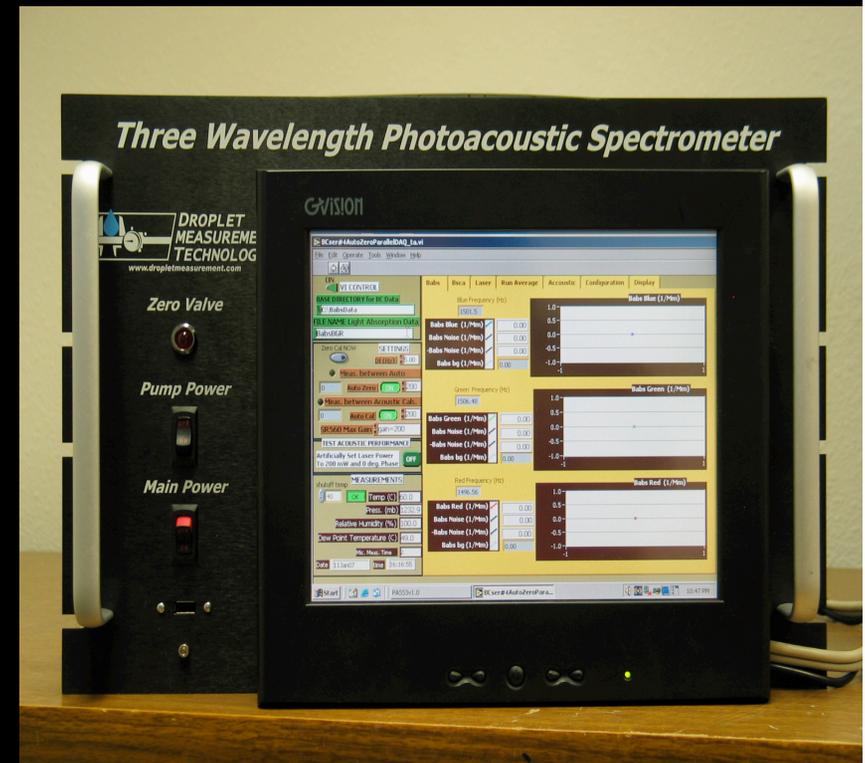
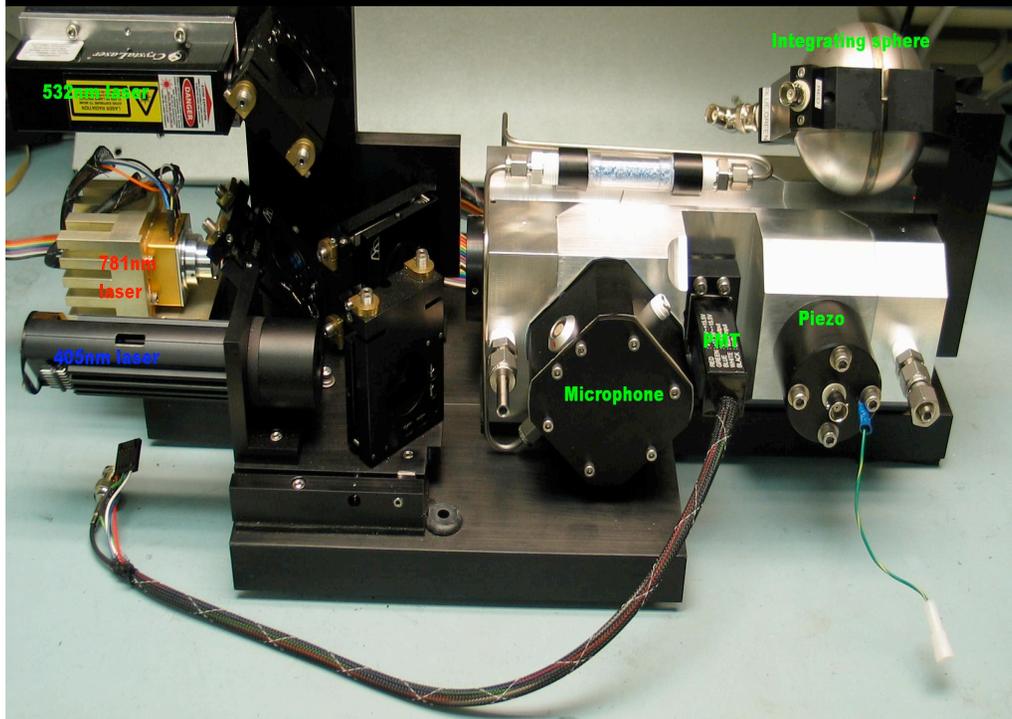
Quinn ACP 2008

Hansen CD 2007

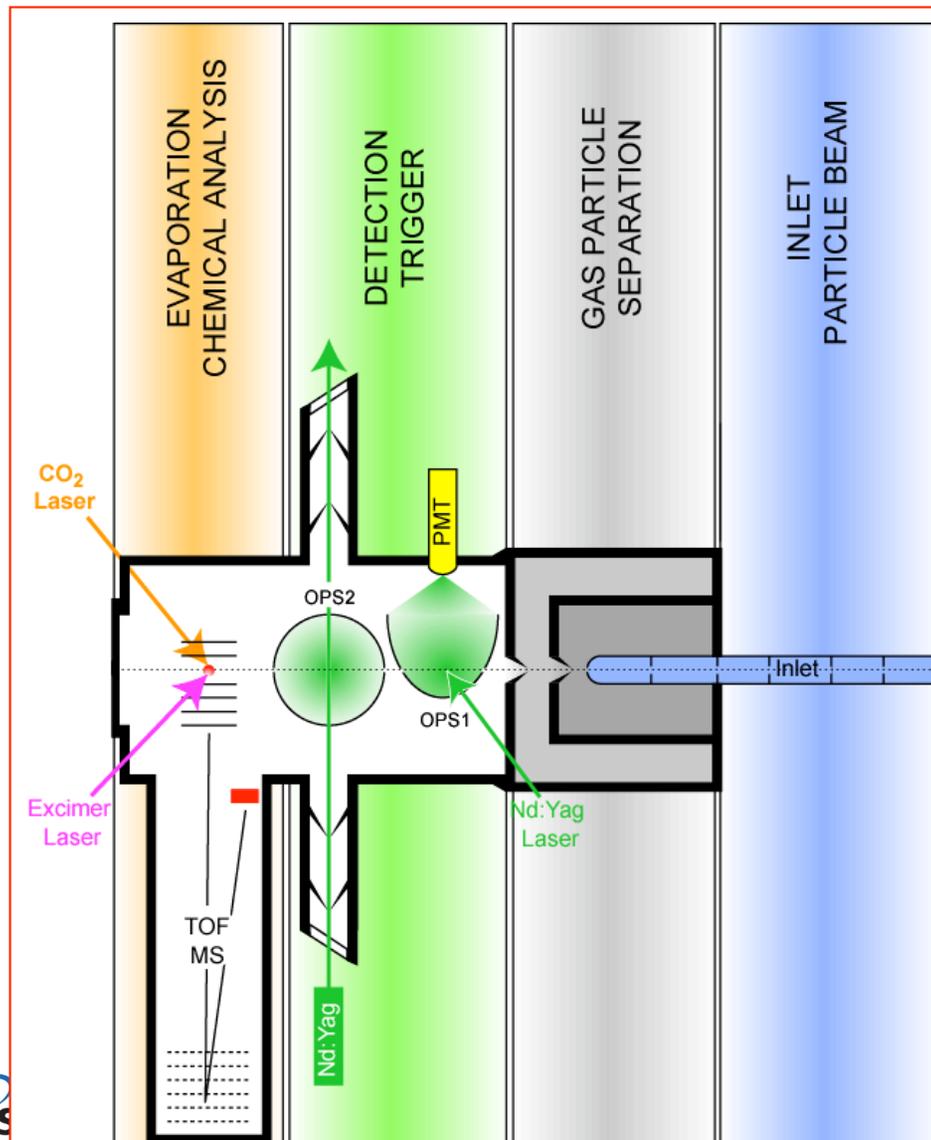


Los Alamos 3-Laser Photoacoustic (DMT) Absorption and Scattering 405, 532, 781nm

- Measures absorption, scattering and single scatter albedo (=scattering/extinction)
- Can discriminate soot, dust and sulfate.
- How dark (warming) or light (cooling) are aerosols?
- What aerosols are good ice nuclei (dust, organics)?
- Do cloud processes darken aerosols as they deposit on snow?



SPLAT II: An Ultra-Sensitive, High-Precision Single Particle Mass Spectrometer (PNNL)



- Provides in *Real-time* the size and internal composition of individual 50 nm to 3 μm particles
- High sensitivity: detects 1p/sec for an aerosol sample of $1\text{p}/\text{cm}^3$ with $d > 125\text{ nm}$
- High sensitivity to small particles: detects 40% of 100 nm particles
- Sampling rate: sizes up to 500 p/sec, 100 of which are also chemically characterized
- Measures refractory and non-refractory aerosol fractions in each particle
- Measures aerodynamic size with better than 1% accuracy

Satellite Retrievals

Global Assimilation

Regional Prediction

Validation

RAQMS

Realtime Air Quality Modeling System

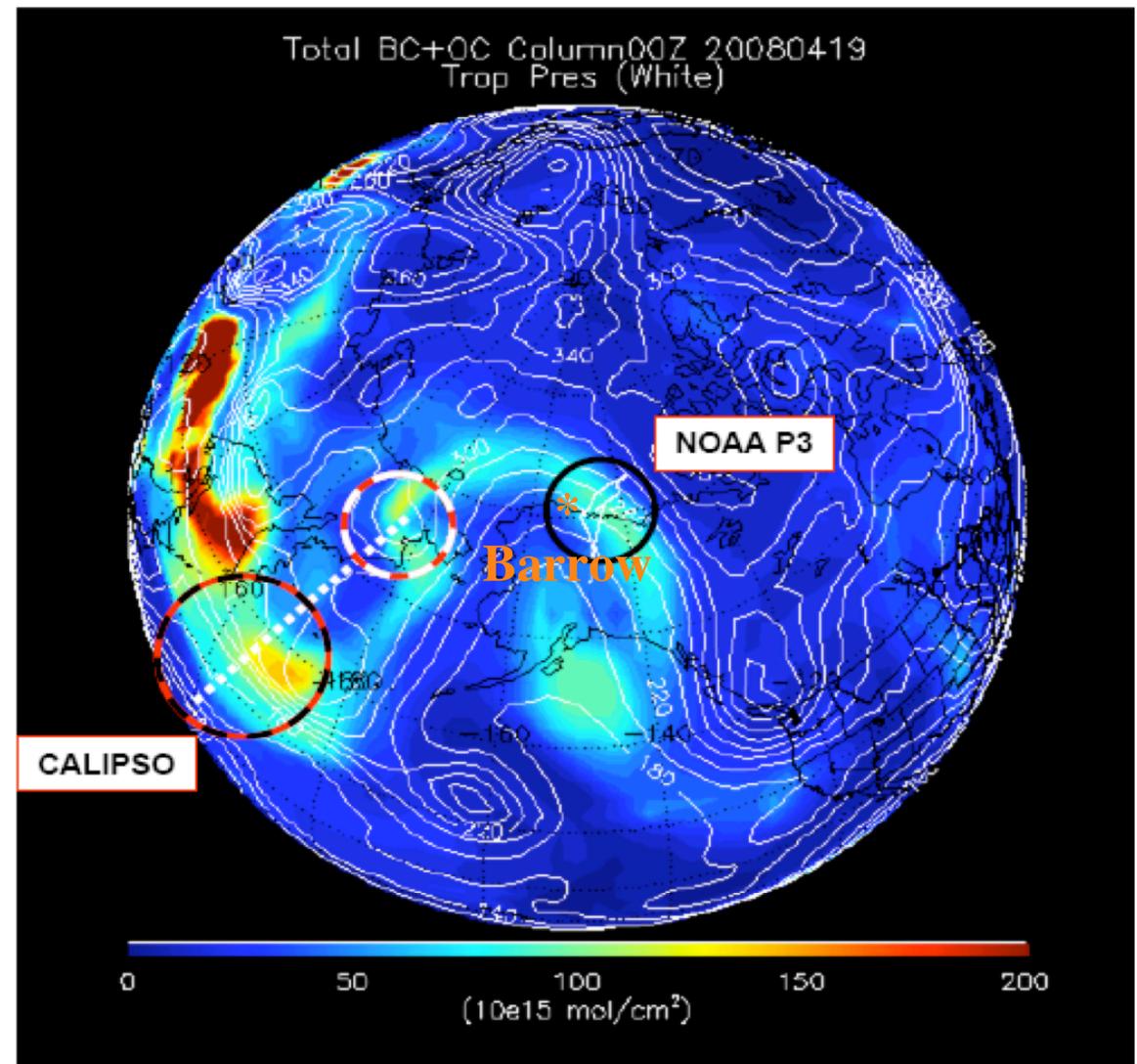
April 19, 2008
BC+OC

RAQMS total column BC+OC (x10¹⁵ mol/cm²) analysis at 00Z on April 19th, 2008.

The tropopause pressure is contoured.

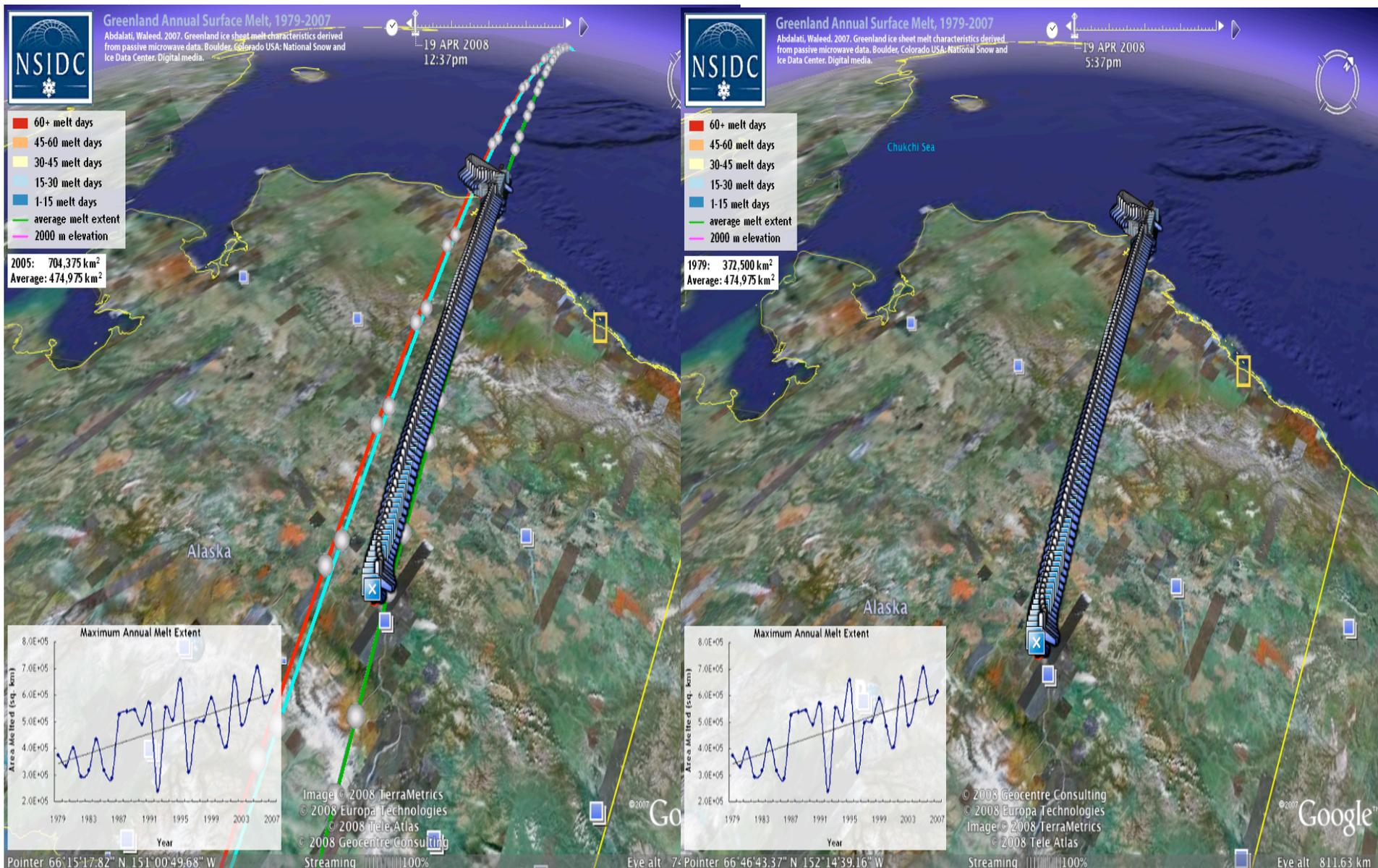
The location of the 15:17:11Z April 18th, 2008 CALIPSO Track is shown as a bold dotted line.

The flight track of the NOAA P3, which sampled the predicted biomass burning plume is also shown off the Northern coast of Alaska.



Brad Pierce, NOAA

C-580 Flight: 19th April 2009, 11.30am – 8.30pm



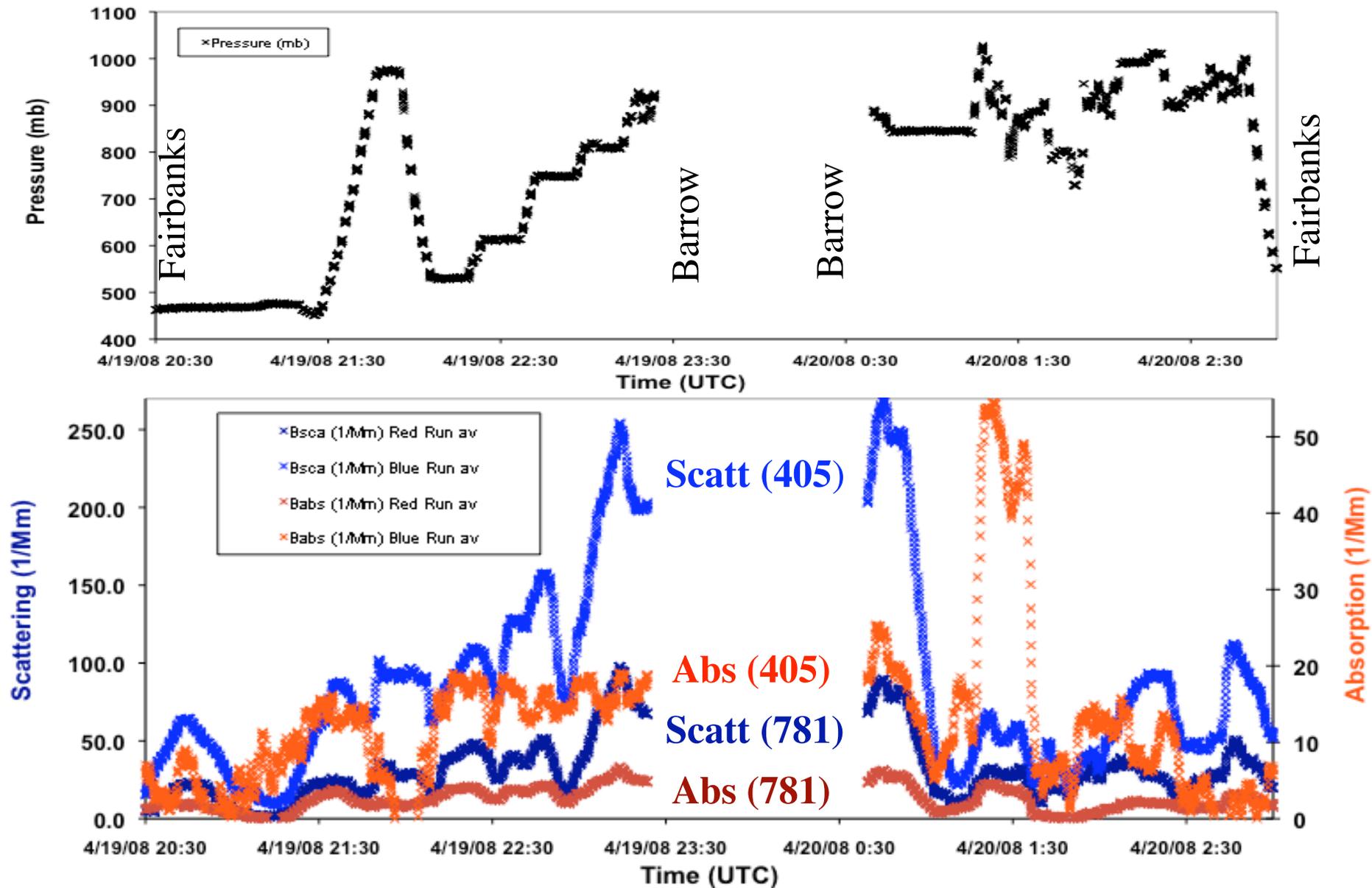
Large Pollution in Polar Regions

(ISDAC 19 April 2008)

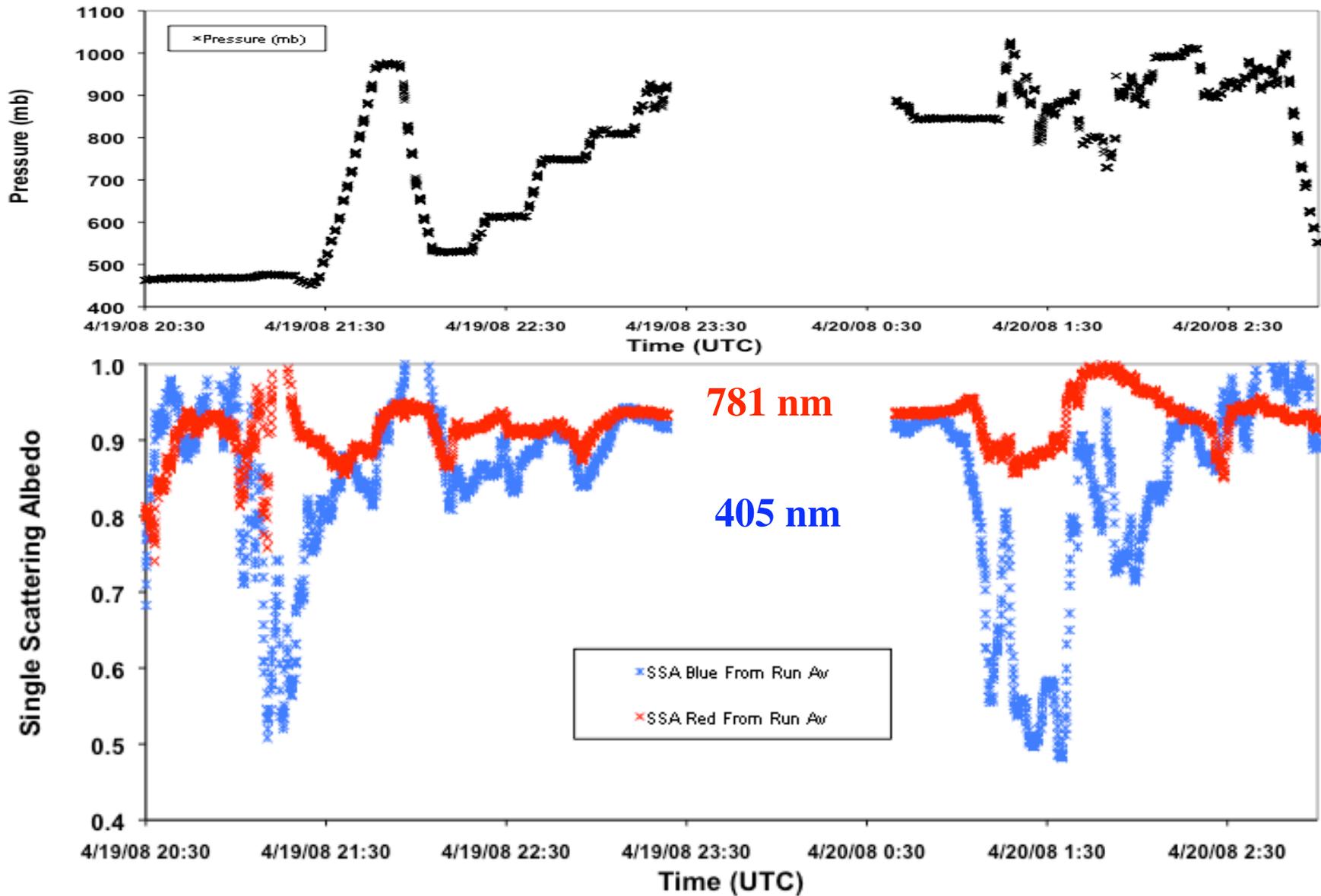
Layer of
Arctic Haze



Flight Track Optical Properties: Time Series

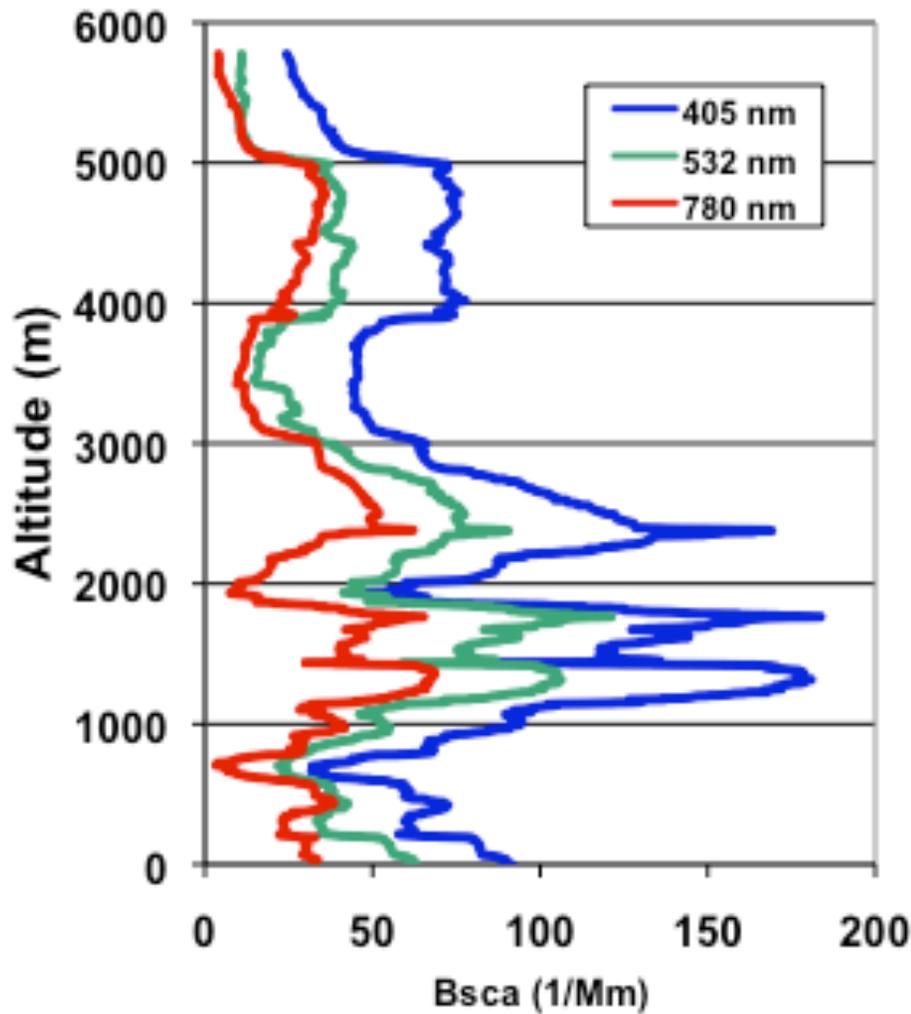


Flight Track Single Scatter Albedo: Time Series

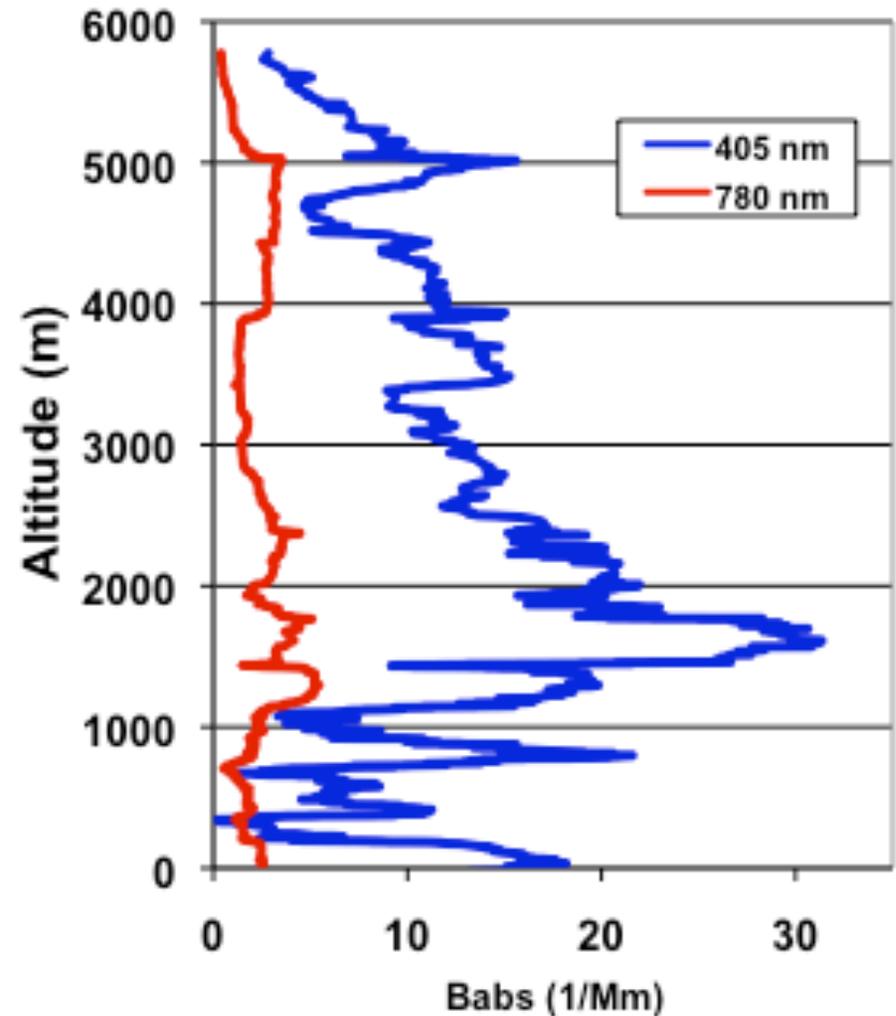


Pollution Layers: Soot, OC, Dust, Sulfate

Scattering Mm^{-1}



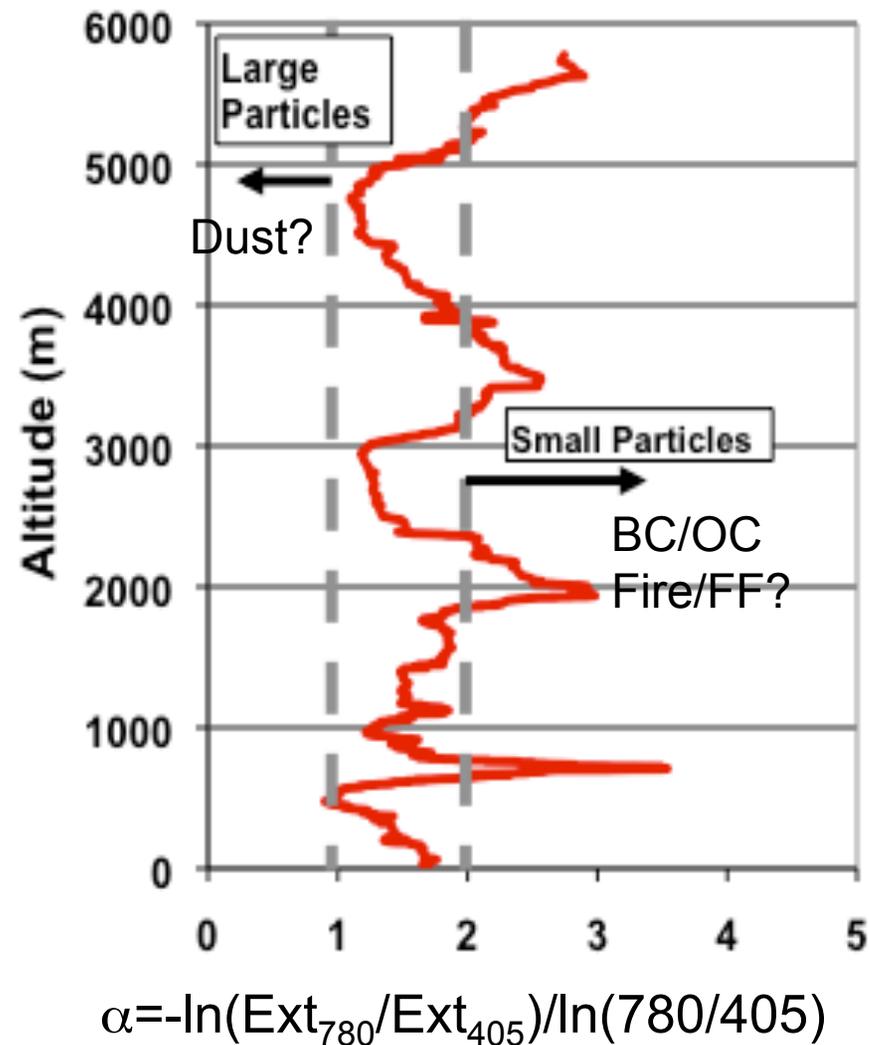
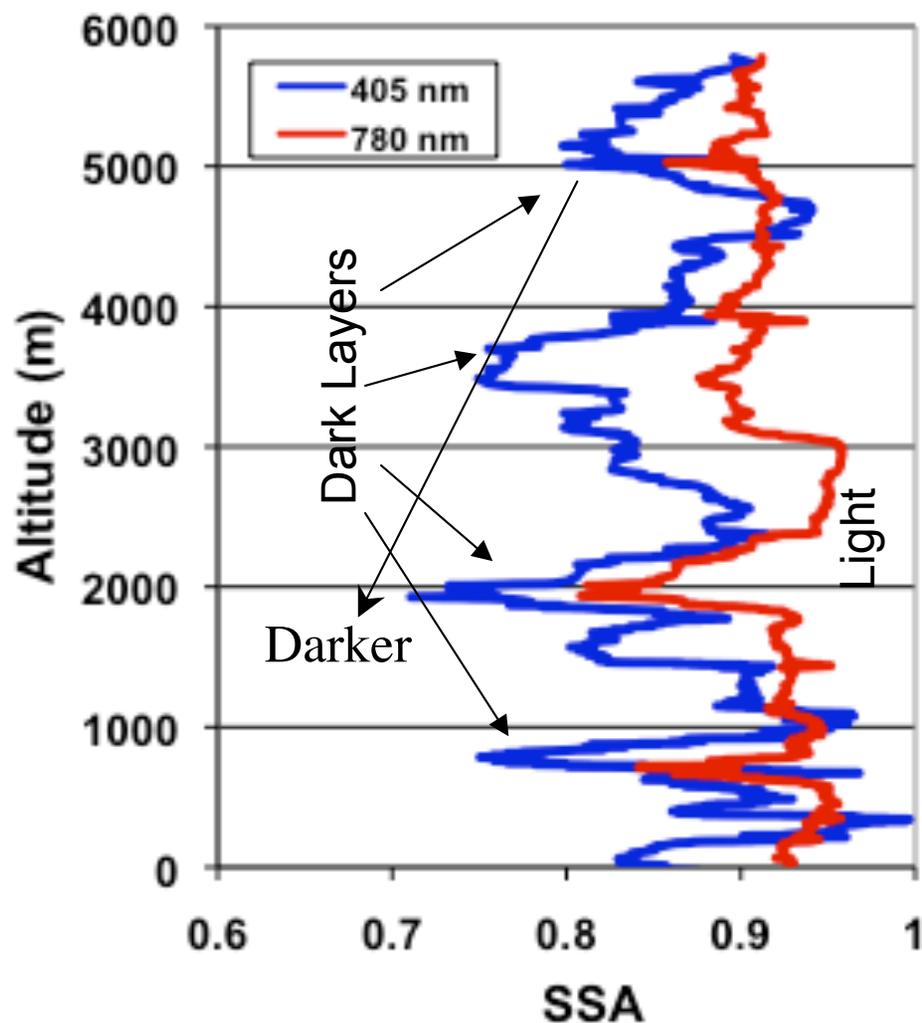
Absorption Mm^{-1}



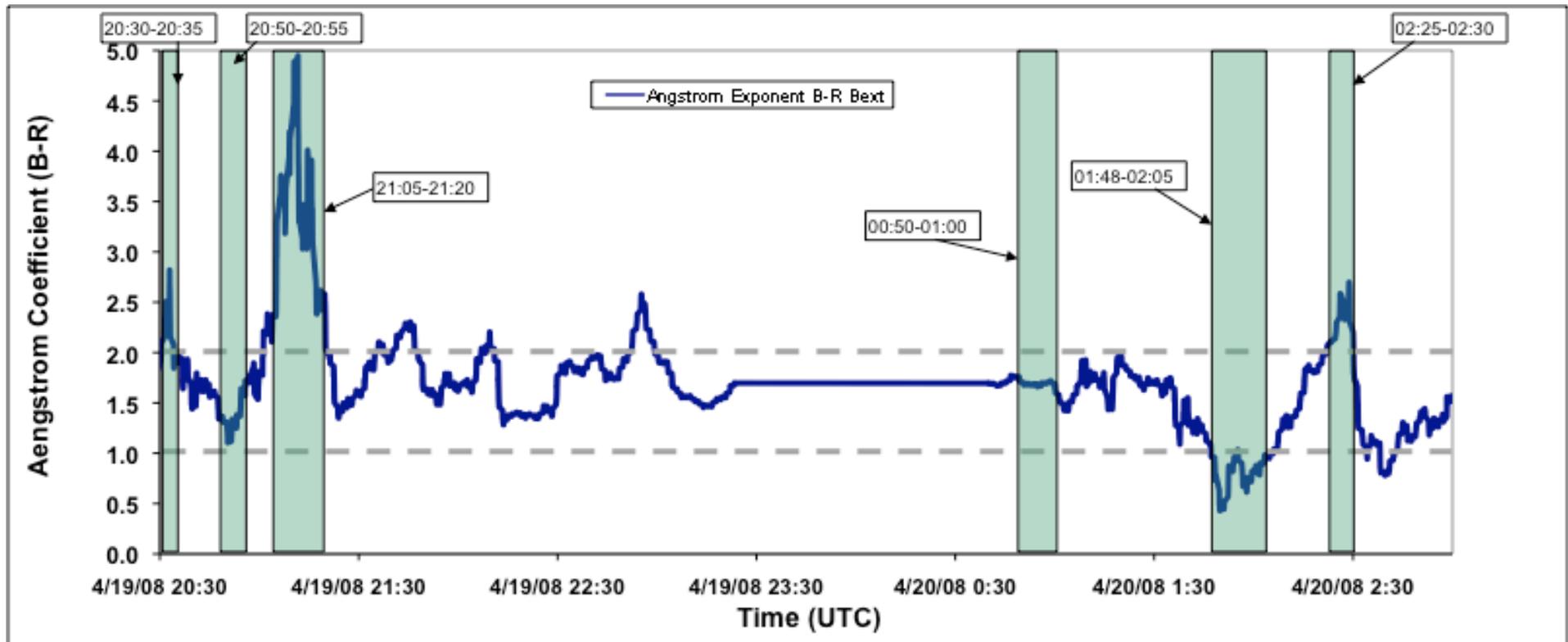
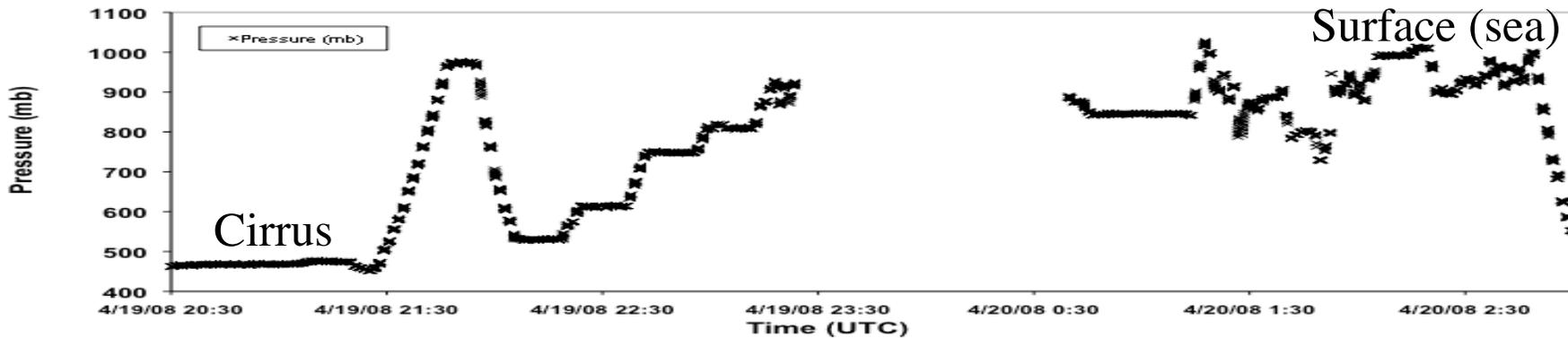
Pollution Layers: Soot, OC, Dust, Sulfate

Single Scatter Albedo [S/(S+A)]

Angstrom Coeffn

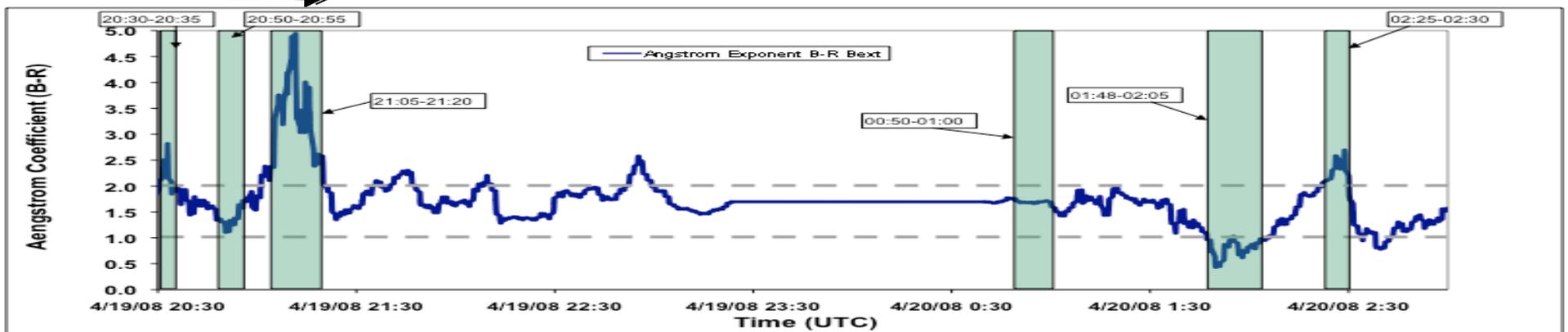
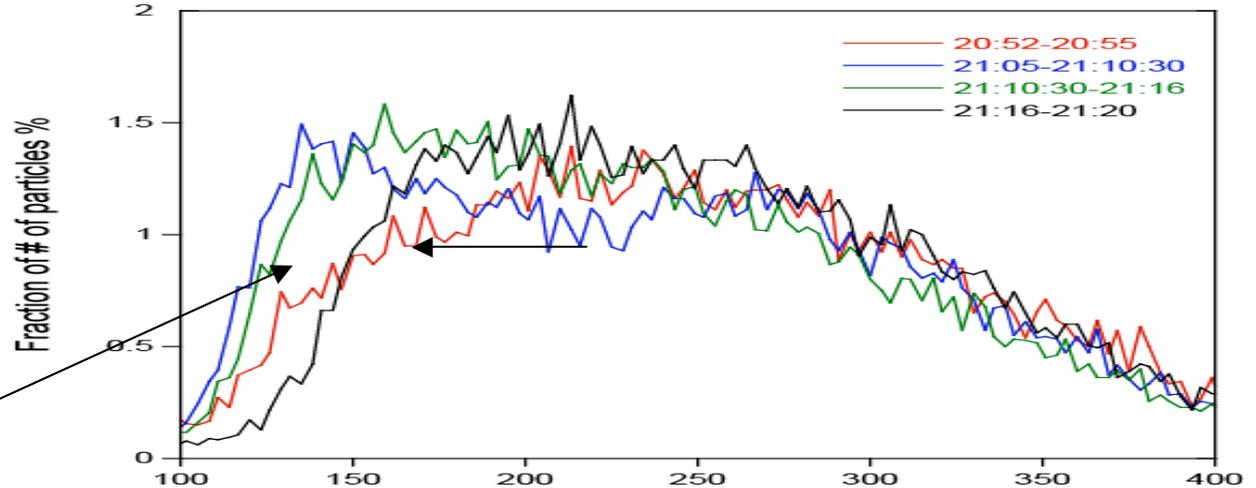
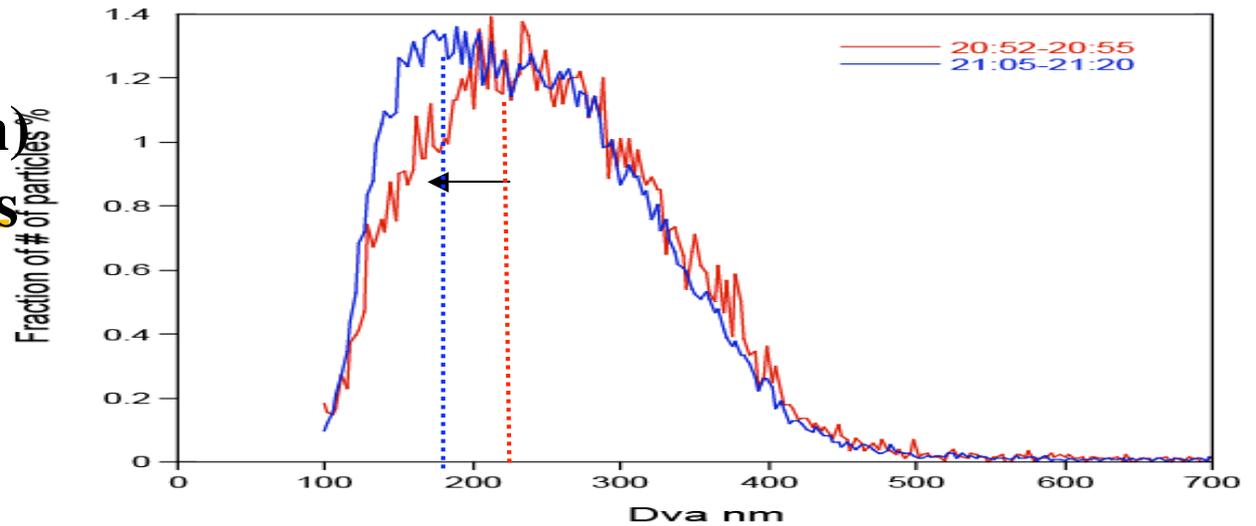


Large Angstrom Exponent Gradients Along Flight Track: April 19

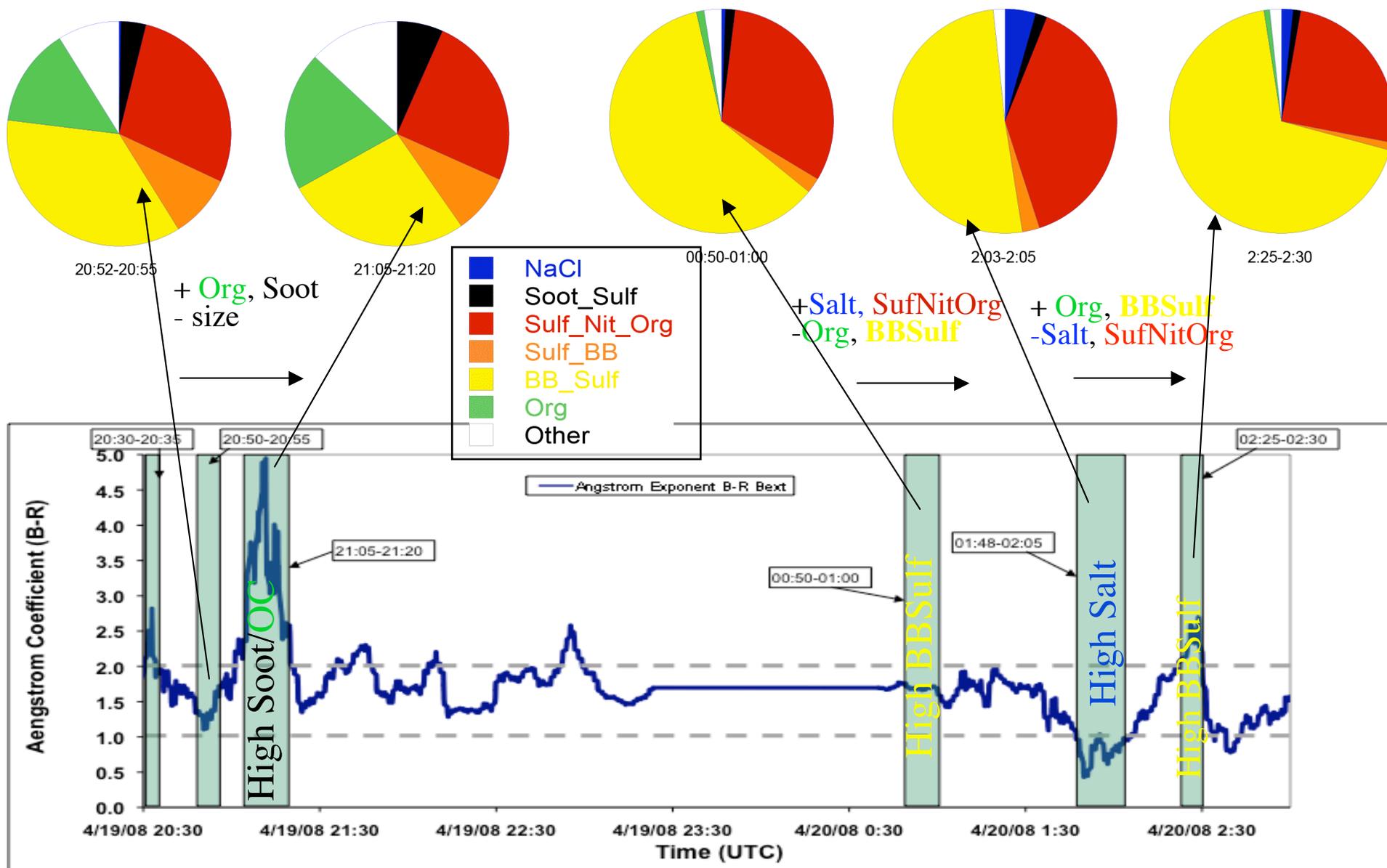


Gradients in Optical Properties (Angstrom) and Size Distributions

During 21:05-21:20 period when Angstrom exponent increased dramatically (and varied) there were more small particles and size distribution was changing quite rapidly with time

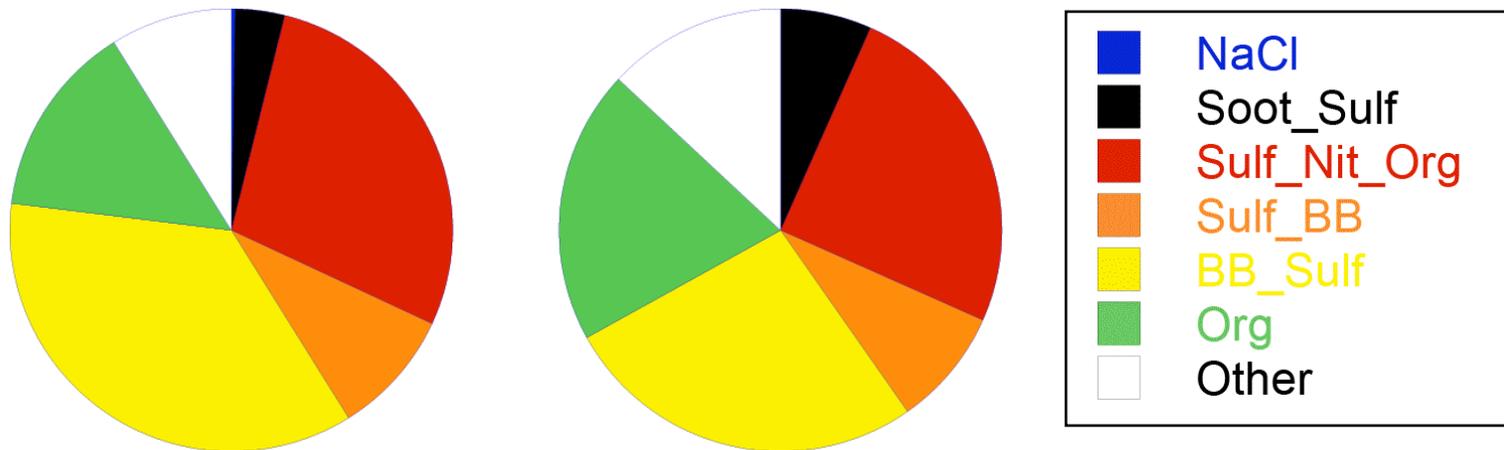


Link Optical Properties (Angstrom Gradients) to Chemical Composition Changes



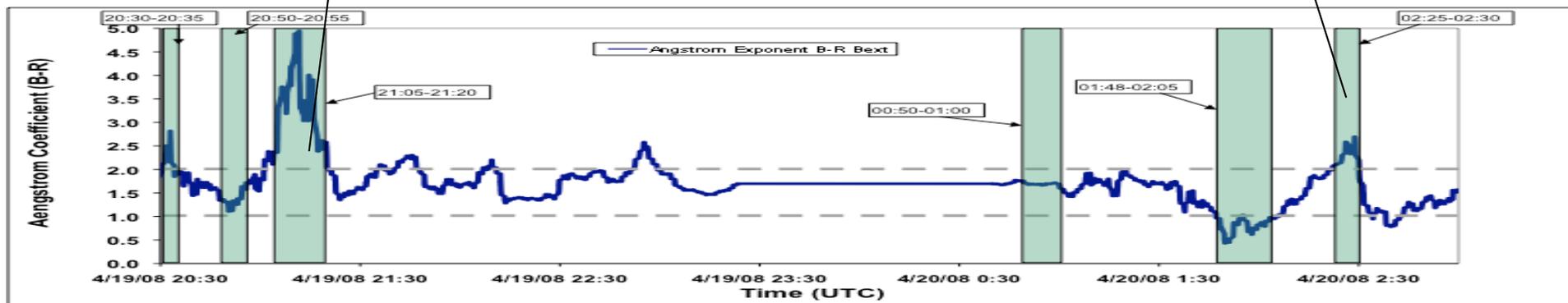
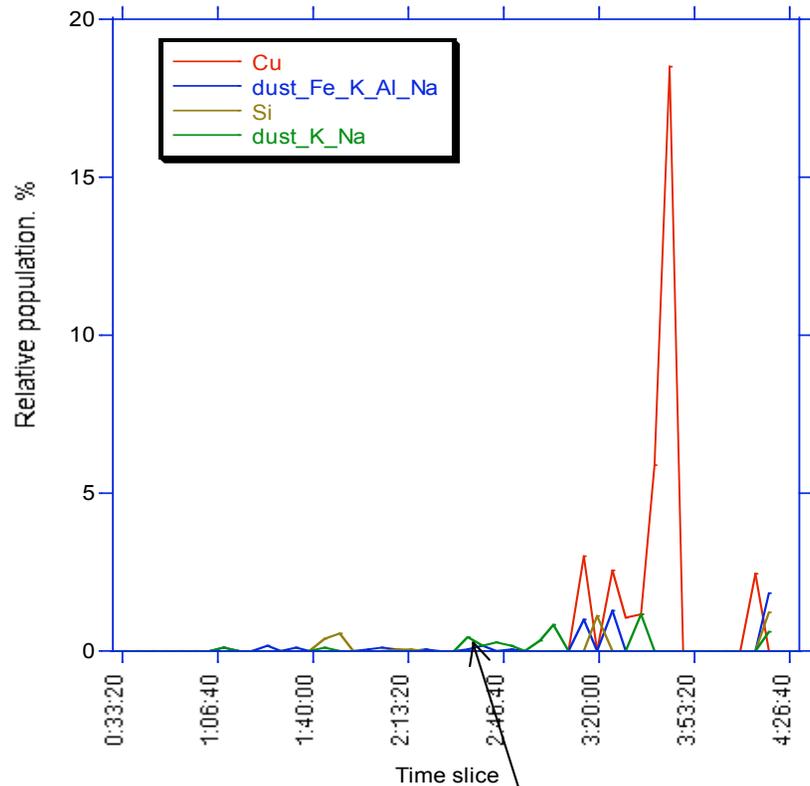
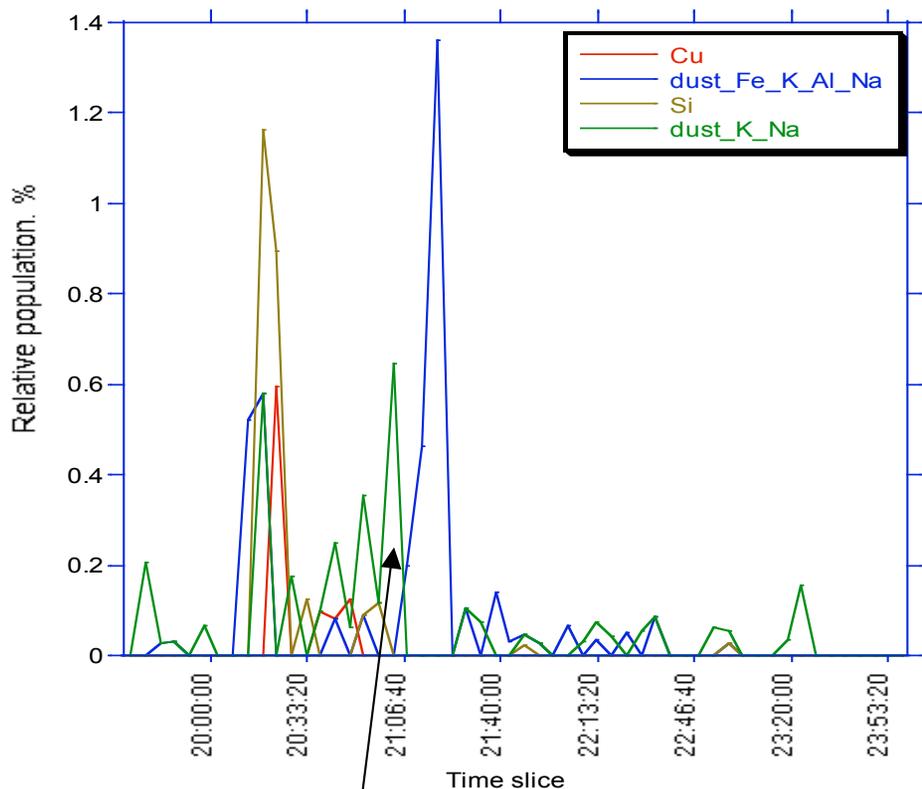
Organic/soot enhancement correlates with smaller particles that are darker in the blue (405 nm)

Soot (~80%) internally mixed with sulfate (20%)
 BB internally mixed with sulfate and vice versa



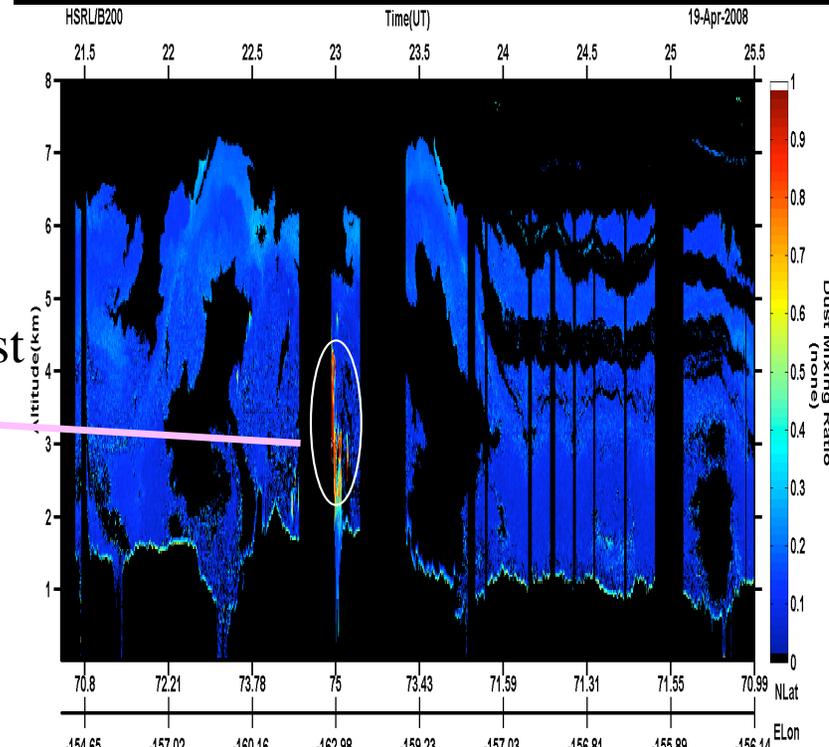
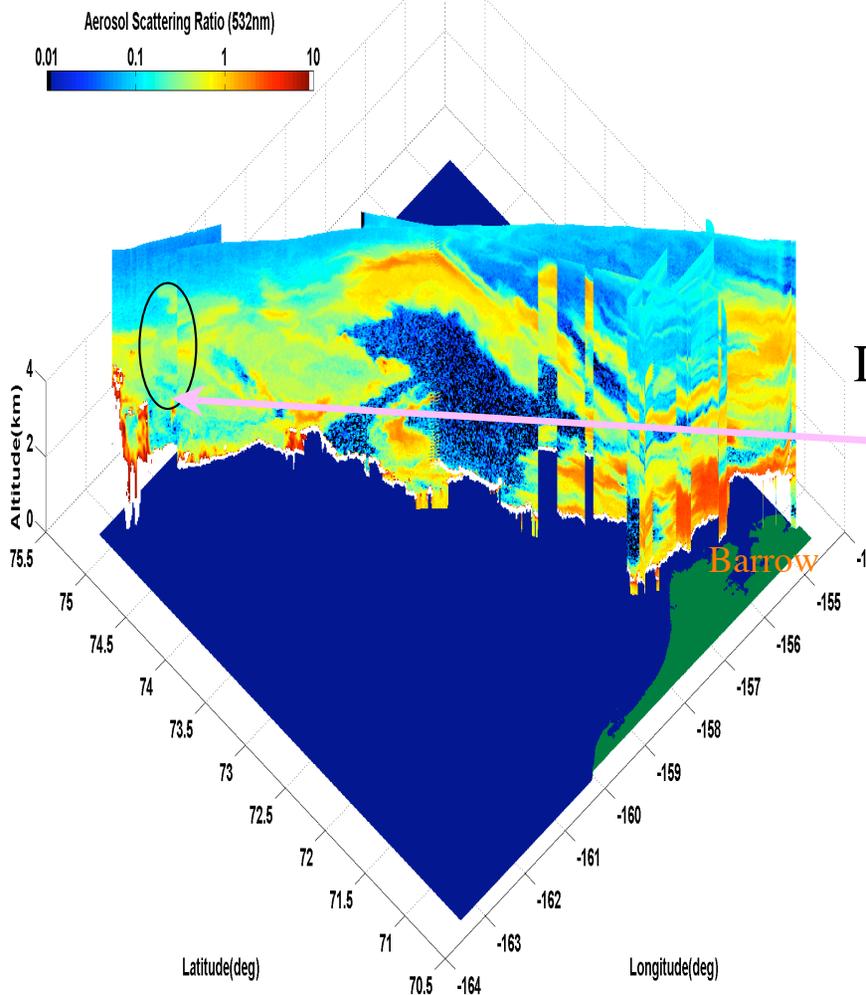
- Increase in Soot & Organic Components (x2)
- Mean diameter decreases from 220nm to 175nm
- Angstrom exponent increases by 2.5
- 405 nm absorption increases x 4, scattering decreases x 1/4
- SSA drops from 0.95 to 0.65

SPLAT observed dust at high altitude but number concentrations were low (1-2%)



4/19 Pollution NASA HSRL/B-200 Lidar Profiles: Depolarization Derived Dust

532nm Scattering Ratio



Dust Fraction from Depolarization Ratio 532 nm

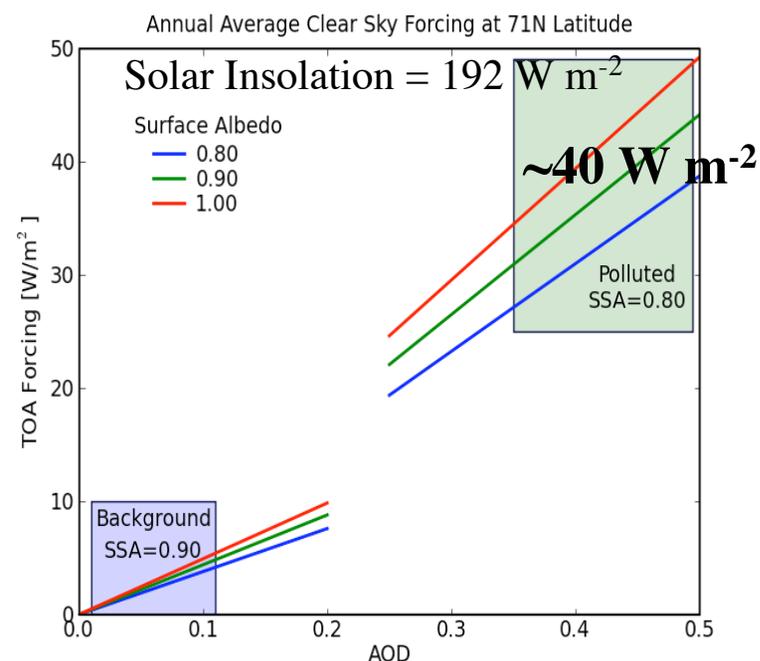
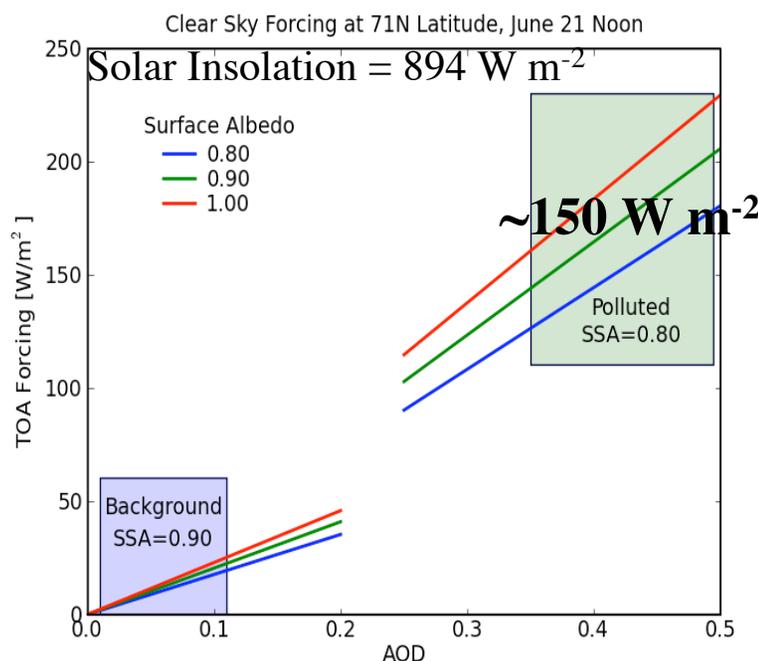
Scale: Ground (Barrow), in situ profile, LIDAR, satellite



UNCLASSIFIED What is clear sky TOA radiative forcing of Arctic Pollution?

S = Solar constant
 T = Transmission
 N = Cloudiness ~ 1
 g = Asymm Par. ~ 0.7
 (AERONET)
 w = Single Scatt. Albedo ~ 0.8
 a = Surface albedo
 τ = Aerosol Optical Depth
 • Chylek GRL, 1995

$$\Delta F = -\frac{S}{4} T^2 (1 - N) \tau [(1 - a)^2 (1 - g) \omega - 4 a (1 - \omega)]$$



Perturbation $\Delta \text{GISS}^{\text{Mod}}$ is 0.92 W m^{-2} (Spr.), 0.3 W m^{-2} (Ann.) Quinn 08, $< 1\%$ of our observation



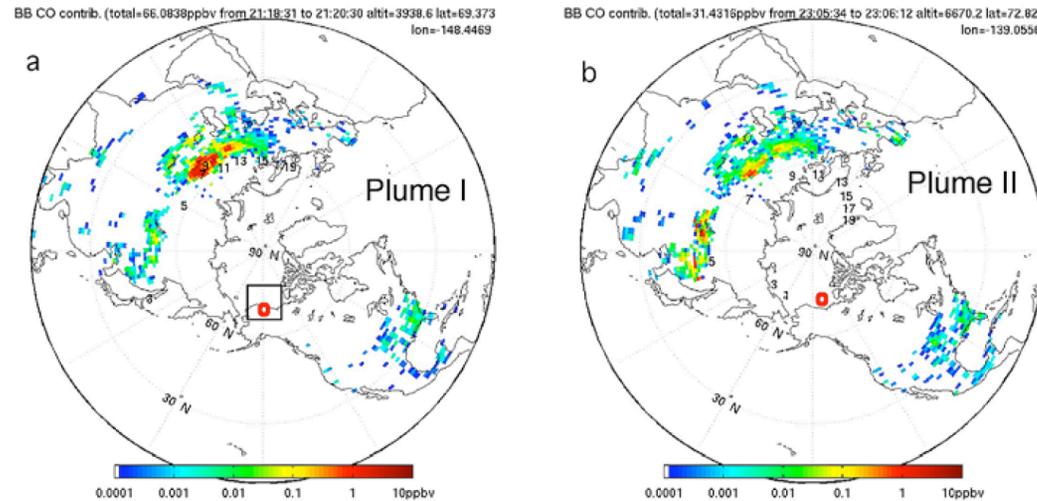
Pollution (Fires, dust, industrial) events cause transient direct forcings that can last for 10-20 days and are orders of magnitude larger than the mean Arctic aerosol and GHG forcing and can be highly variable.



Biomass burning in Siberia and Kazakhstan as an important source for haze over the Alaskan Arctic in April 2008

C. Warneke,^{1,2} R. Bahreini,^{1,2} J. Brioude,^{1,2} C. A. Brock,¹ J. A. de Gouw,^{1,2} D. W. Fahey,¹ K. D. Froyd,^{1,2} J. S. Holloway,^{1,2} A. Middlebrook,¹ L. Miller,^{2,3} S. Montzka,³ D. M. Murphy,¹ J. Peischl,^{1,2} T. B. Ryerson,¹ J. P. Schwarz,^{1,2} J. R. Spackman,^{1,2} and P. Veres^{1,2}

BB CO Plume
Fires, Flexpart



MODIS fire

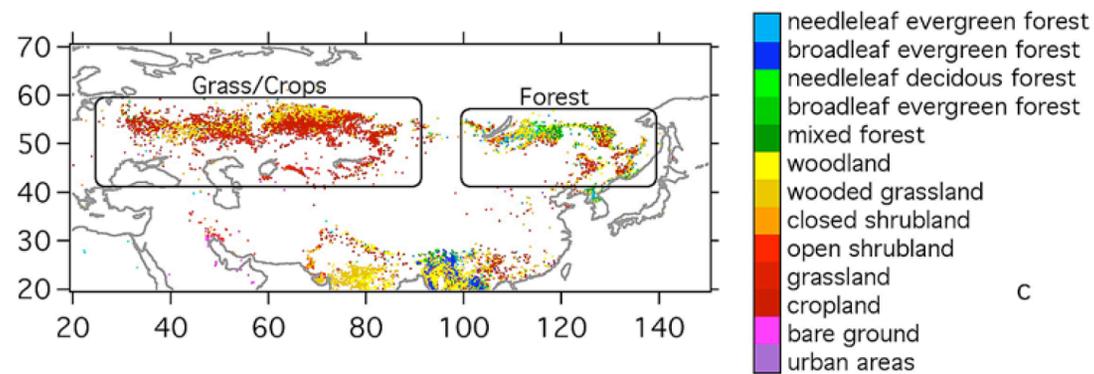


Figure 2. (a) and (b) FLEXPART BB CO source contribution for plume I and II in Figure 1 on a logarithmic scale. The red circle in Figure 2a indicates the area where the ARCPAC flights took place and all the BB plumes were observed. The color code indicates the dominant land cover type for each fire detection between April 1, 2008 and April 20, 2008.

Conclusions

- Arctic haze optical properties consistent with layers of aged biomass-fires, dust and sulfate
- Internally mixed Soot-Sulf, Sulf-BB, Sulf-Nit-Org
- Optical property gradients correlated to changes in chemical composition and size distributions: Needs Closure Studies
- Our SSA imply large episodic forcing above Arctic in April that are not captured in current climate change projection models

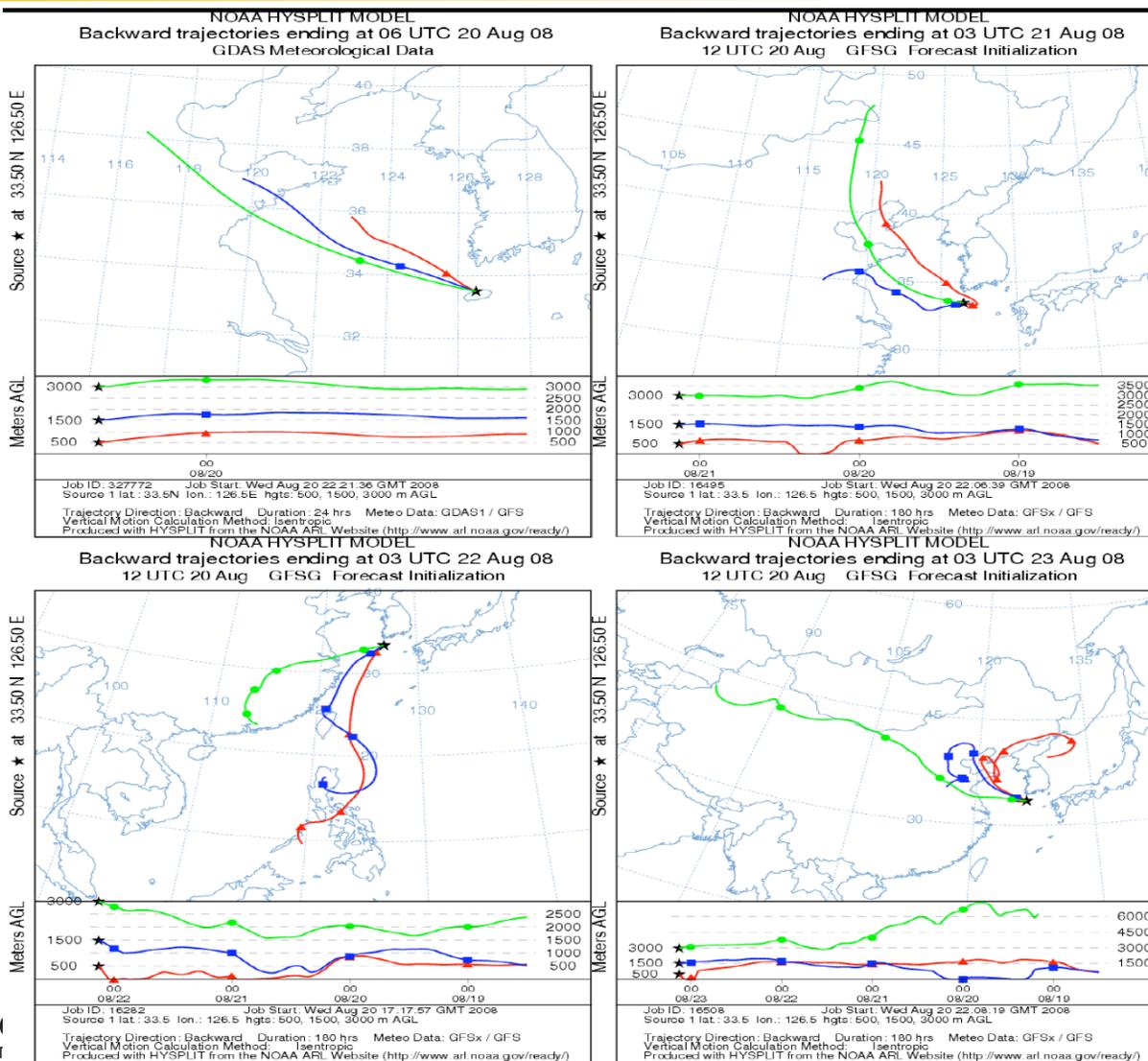
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Photoacoustic Observations of Aerosol Optical Properties *at Cheju Island Korea*

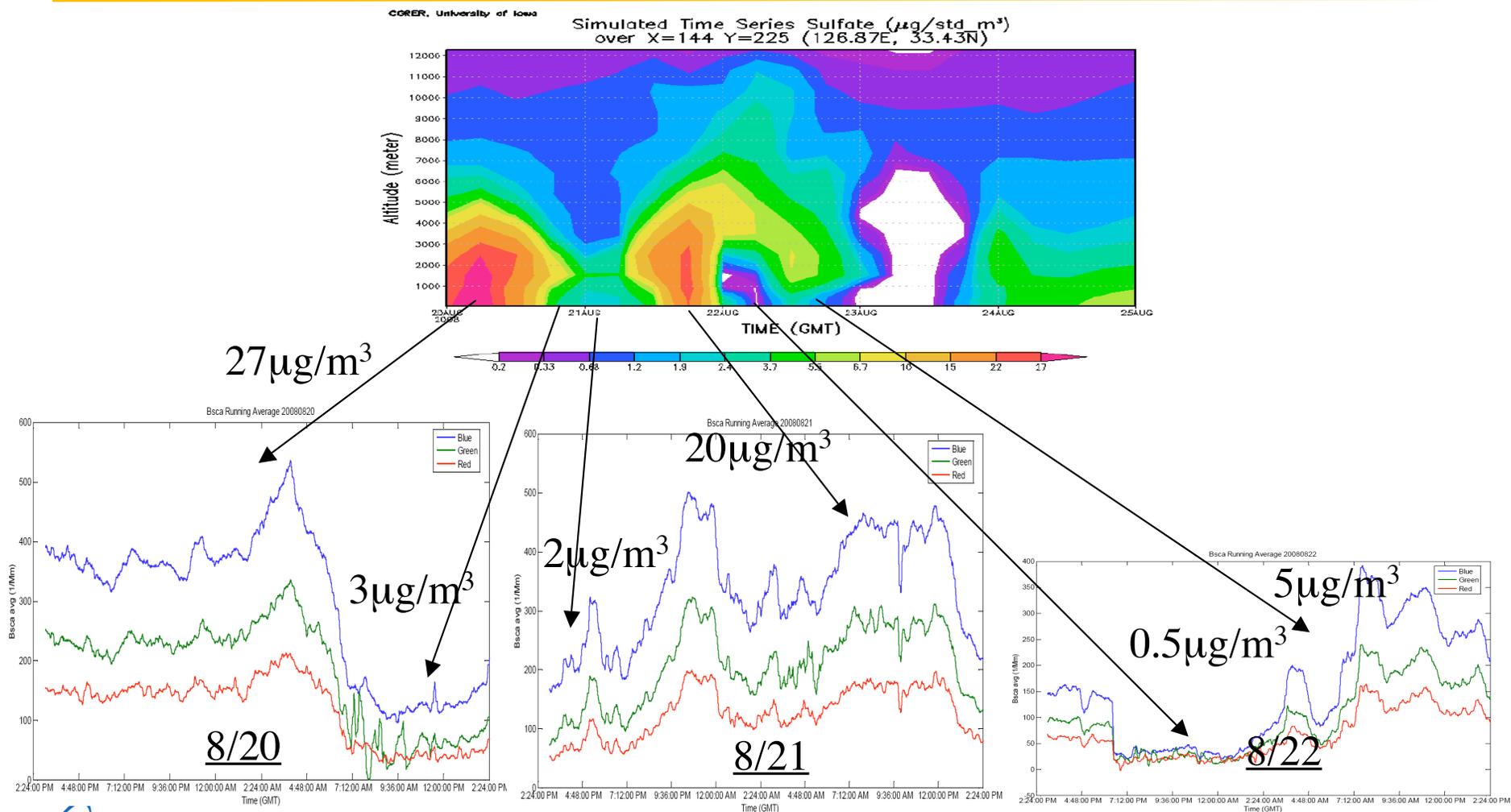
*Manvendra Dubey (LANL), Claudio Mazzoleni
(MTU, LANL), Alicia Garcia Lopez (LANL) V.
Ramanathan, the CAPMEX Team (Beijing
Olympics Aug-Sept 2008)*



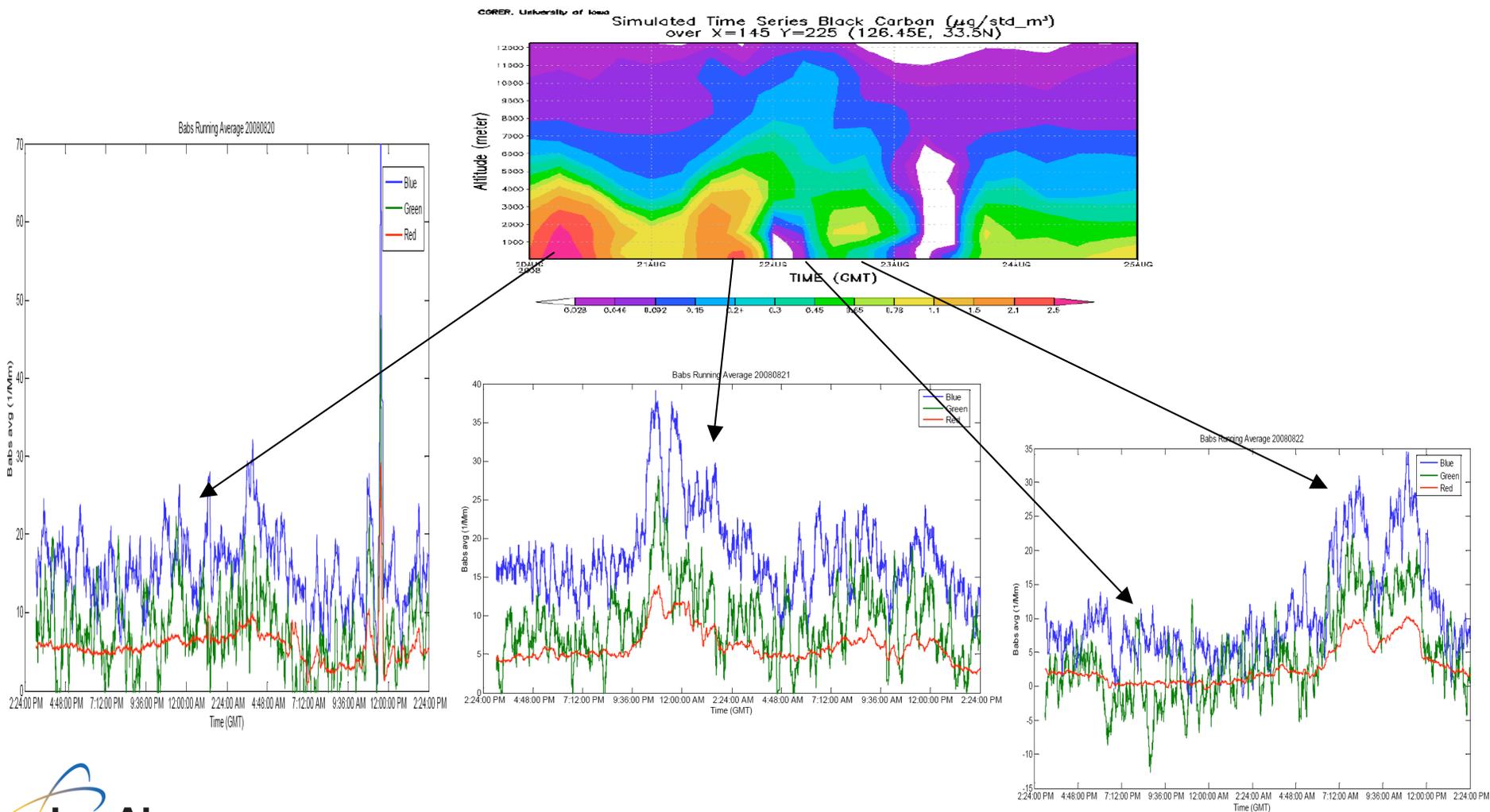
Back-trajectories 8/20-8/23: Changing source regions



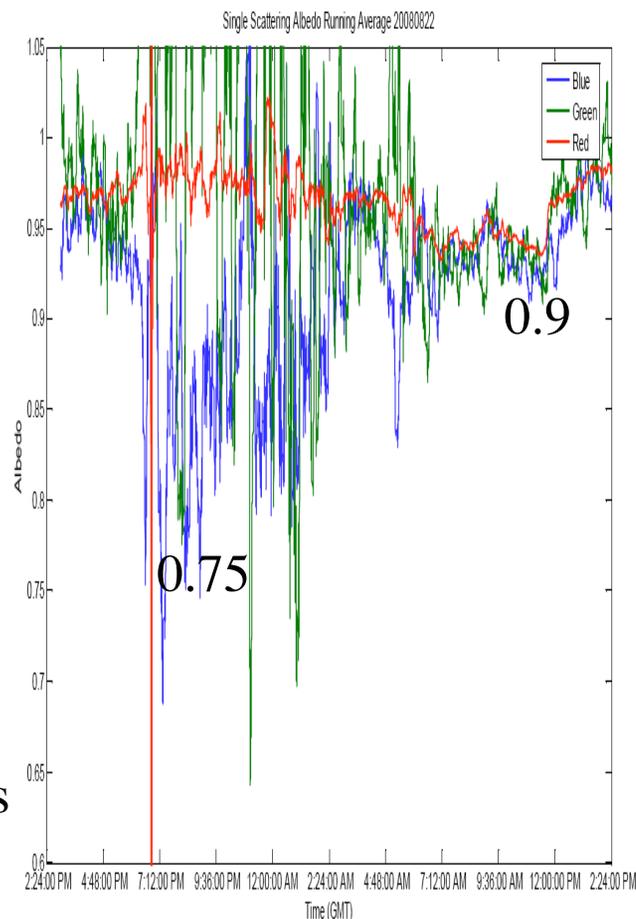
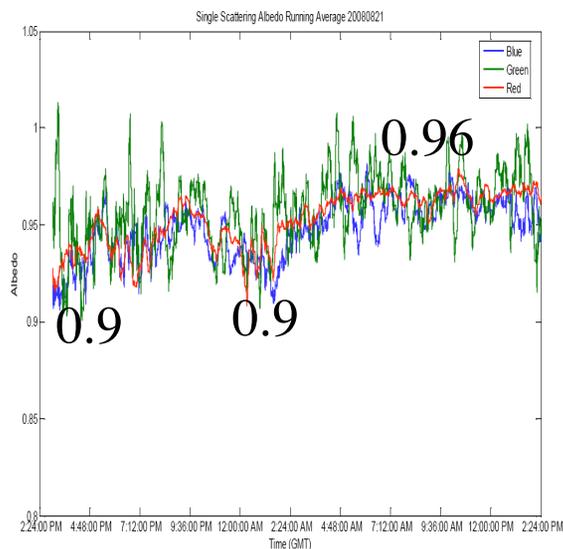
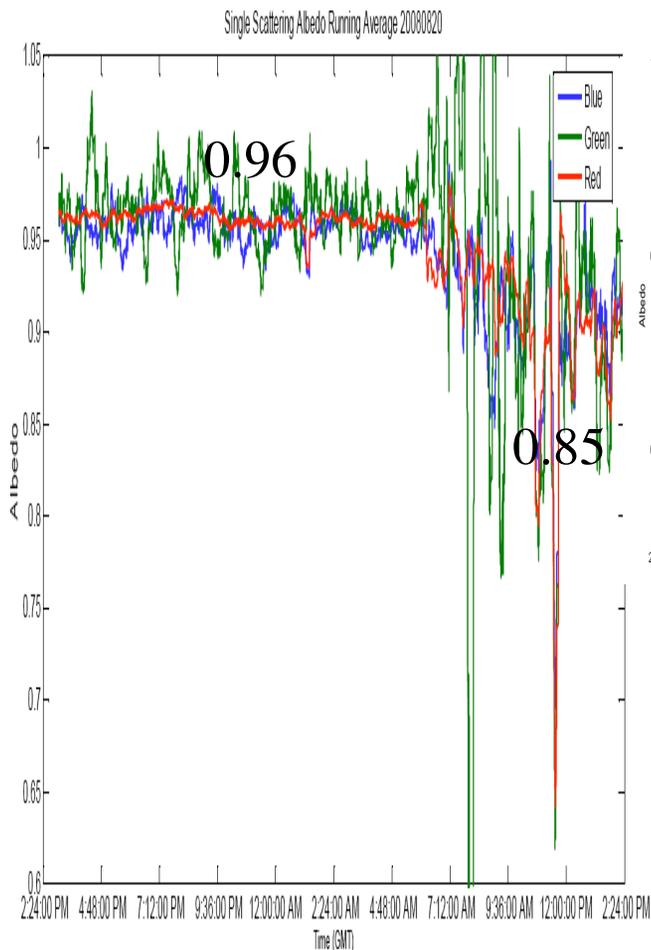
Observed Scattering and Modeled Sulfate: 8/20-8/22



Observed Absorption and Modeled Black Carbon: 8/20-8/22



Single Scatter Albedo 8/20-8/22



Need model predictions using composition and mixing state assumptions to test reality!